Uranium and thorium in Archean granulite facies terrains of Bahia (Brazil)

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Th and U distributions in Archean granulite-facies rocks from Bahia (Brazil) have been investigated by means of gamma-ray spectrometry. Thorium shows a very large range of values (from <0.3 to 72.2ppm) with high concentrations in only a few samples. Uranium is more homogeneously distributed at abundance levels below 1ppm. The Th/U ratios vary irregularly depending on the variable Th contents but are normally much higher than averages for magmatic and metamorphic rocks or crustal averages. Differential behaviour of Th and U during metamorphism is considered as the main cause of the high Th/U values observed. Selective depletion of uranium in metamorphic fluids may have resulted from an early event as documented by other very old crustal materials. Surface heat production data indicate that the Bahia granulitic block is a low-heat production region (average H.P.U. 3.32). Emphasis is placed on the need for a future study of the vertical compositional profile of the crust in shield areas with exposed high-grade metamorphic rocks using combined heat production rate and heat flow data.

INTRODUCTION

Available heat production data for igneous rocks define the well known igneous trend characterized by a marked increase of radioelements with differentiation. However, heat production in metamorphic rocks has not received comparable attention, partly because estimates of average heat production in the various rock types are complicated by the strong compositional heterogeneity of the metamorphic terrains related both to the premetamorphic nature of the rocks and to metamorphic effects. New models for continental crust, taken as a basis for geothermal calculations, emphasize the importance of metamorphic rock volumes as the main components. For example, the model of SMITHSON and DECKER (1974) suggests a three-fold layering of the crust with the lowest and the thickest (about 18km) layer consisting of amphibolite-granulite facies rocks. Heat production data for these rocks are therefore of primary importance in any large-scale geothermal calculations, for example in the interpretation of thermal flow data in stable continental regions.

This paper presents the results of a preliminary investigation on the distribution of the radioelements in Archean granulite facies terrains in northeastern Brazil. From the analyses carried out in the complete program it was hoped to determine: a) the average uranium and thorium content in this area of the crust, b) the relationship of U and Th abundances to other chemical and petrologic variables using deductions on starting materials for granulites and the effects of metamorphism. In addition, by comparison of surface heat production data with heat flow data, we hope to obtain information on the composition of the earth's crust in deeply eroded shield areas.

GENERALITIES ON THE GRANULITE FACIES TERRAINS

Data on granulitic rocks from Bahia are reported in numerous papers (e.g., FUJIMORI, 1968; SIGHINOLFI, 1970, 1971). Granulitic rocks recently interpreted by FYFE and LEONARDOS (1974) as a charnockitic belt of dominant low-pressure type constitute the most common rock type in Bahia State, in particular in the eastern

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part, where they are well exposed in N-S trending belts. In Eastern Bahia granulites are well preserved and relatively little affected by post-orogenic processes; in Western Bahia, however, they are strongly affected by retrometamorphism and migmatization. Age data for most of the granulitic terrains give approx. 2.0×10^9y which corresponds to the Transamazonic event (CORDANI, 1973). However, more recent data, especially on rocks from areas of Central Bahia (Jequié complex), reveal the presence of much older rocks (*ca.* 3×10^9y). Data of the present paper refer mainly to granulite samples coming from an area of Central Bahia situated between the localities of Arguim, Iaçú and Itaberaba. Here granulites, constituting almost totally the basement, contain komatiitic amphibolites typical of Archean volcanic sequences and small bodies of pyroxenitic ultramafics (ROCHA, 1975). To the west (near Itaberaba) superimposed migmatization has strongly affected granulitic rocks which occur as remnant bodies of a few meters in size. Systematic sampling was carried out on the road Arguim-Itaberaba in a W-E direction, i.e. cutting perpendicularly the main structures of the area. Samples were chosen for analysis after rigorously checking that they are representative of the most diffused rock types. Granulitic rocks from other areas of Bahia State have been also analyzed to provide preliminary information on other fragments of the granulitic block (Table 1). Forthcoming papers will present further data on Th and U distributions in granulitic areas listed in the table.

ANALYTICAL NOTES

Detailed data on the analytical method used were previously reported (SIGHINOLFI and SAKAI, 1974). Uranium and thorium were determined by gamma-ray spectrometry, using a 1024-multichannel analyzer (5401 B Hewlett-Packard mod.) connected to a 10 cm × 8 cm NaI (T1) crystal. Using 800g of crushed rock sample, Th was determined from the 208 T1 peak (2.62MeV); U from 215 Bi peak (0.607MeV), after subtraction of the 208 T1 contribution by the tangent method. The maximum error is estimated to be about 10% for Th and 20% for U.

ANALYTICAL RESULTS

The results of the analyses for Th, U and K as well as the Th/U ratios and heat production data are given in Table 1. The limit of sensitivity for thorium is about 0.3 ppm and that for uranium depends on the absolute Th content. For purposes of calculation, the Th and U contents of a few samples in which they are below the determination limit are, rather arbitrarily considered to be a half of this value. Radioelement partial averages were calculated for the Itaberaba area which was studied in detail. These data show that for thorium there is a large range of values ($< 0.3 - 72.2$ ppm) even for samples collected at short intervals. For this reason and also because of the limited number of samples analyzed the calculated Th averages are rather meaningless and comparison with other Th averages are quite insignificant. On the contrary the uranium content of almost all the samples is less than 1 ppm. The U average for the 31 samples analyzed is 0.70 ppm; this value would be strongly reduced (0.51 ppm) by excluding sample RP 4, which is anomalously enriched in U. The limited number of samples analyzed precludes a quantitative study of the distribution statistics. However, qualitative observations indicate that uranium shows a negatively-skewed frequency distribution with a bias towards values less than 0.4 ppm. The calculated U average is similar to other averages for granulite facies rocks (see e.g. HEIER and ADAMS, 1965; LAMBERT and HEIER, 1968; ROELANDTS, 1973; DEL FIORE *et al.*, 1974; KILLEEN and HEIER, 1975a) but is markedly lower than the average for Canadian granulites (4.16 ppm, BURWASH and CUMMING, 1976) as well as being lower than the average for the entire Canadian shield calculated by data on composite samples (SHAW, 1967; EADE and FAHRIG, 1971).

Thorium averages 13.34 ppm in the Bahia granulites. Notwithstanding the large range of values, these rocks are relatively rich in thorium when compared with other high-grade metamorphic rock types (Table 2). The relative concentration of thorium with respect to uranium and the large dispersion of the first element is reflected in the Th/U values which in the Bahia granulites are much higher than in other metamorphic rock types or crustal values in general. This peculiar character of the granulites studied will be considered later in the discussion of the metamorphic behaviour of the radioelements.

The heat production data (average value 3.32 H.P.U.) indicate that, essentially because of the low U content, the granulitic block of Bahia represents a low heat-production region

Table 1. Concentrations of radioelements in granulite facies rocks from Bahia.

Sample		Th ppm	U ppm	K(%)	Th/U	H.P.U.
Itaberaba area (Central Bahia)						
RP	1	2.5	0.7	2.92	3.6	1.53
RP	1A	4.9	0.4	4.06	12.2	2.01
RP	2	22.3	<1.0	3.02	>22.3	4.80
RP	3	0.8	0.3	0.71	2.7	0.49
RP	3A	1.2	0.4	0.87	3.0	0.65
RP	3B	45.2	<2.0	4.69	>22.6	9.38
RP	4	72.2	6.3	4.55	11.5	17.23
RP	5	4.0	0.5	3.98	8.0	1.91
RP	6	21.3	<1.0	3.44	>21.3	4.72
RP	7	10.4	0.8	2.35	13.0	2.80
RP	8	33.1	1.3	2.04	25.5	6.90
RP	9	3.8	0.5	1.89	7.6	1.39
RP	10	10.8	0.4	5.02	27.0	3.24
RB	5	20.6	<1.0	4.29	>20.6	4.80
RB	5B	34.9	1.7	3.04	20.5	7.69
RB	53	37.4	<1.5	4.13	>24.9	7.77
RB	78	13.4	0.5	2.07	26.8	3.06
RB	226	7.6	0.3	1.99	25.3	1.94
RB	254	5.9	0.2	2.30	29.5	1.66
Average (19 samples)		18.54	0.92	3.02		4.42
Stand. dev.		18.82	1.35	1.27		4.06
Salvador (Eastern Bahia)						
UB	2	8.0	0.4	4.33	20.0	2.60
UB	10	11.5	0.6	4.42	19.2	3.34
UB	15	<0.3	<0.2	2.02		0.55
UB	16	16.3	0.5	2.42	32.6	3.64
P	136	0.7	0.2	1.93	3.5	0.69
Various localities						
P	127	7.3	0.3	1.20	24.3	1.70
P	129	1.0	<0.2	0.37	>5.0	0.32
p	323	6.1	0.4	1.22	15.2	1.57
CQ	3	4.0	0.3	3.94	13.3	1.77
ES	35	4.5	0.9	1.91	5.0	1.76
Ita	1	<0.3	<0.2	1.21		0.37
IP	48	1.4	0.2	1.73	7.0	0.76
Total av. (31 samples)		13.34	0.70	2.71		3.32
Stand. dev.		16.34	1.10	1.33		3.51

Heat production in units (H.P.U.) of $10^{-13}\cdot cal\cdot cm^{-3}\cdot sec^{-1}$ (assuming specific gravity = $2.67 g/cm^3$). All values computed from given Th, U and K data, using H.P. = 0.62U(ppm) + 0.17Th(ppm) + 0.23K(%) (after KILLEEN and HEIER, 1975a).

of the Brazilian Precambrian analogous to other shield areas, where high-grade metamorphic rocks are exposed.

The distribution of radioelements in metamorphic rocks when compared with that in other rock types may be useful in investigating the nature of the premetamorphic material and/or the possible effects of metamorphism. In magmatic rocks in general both Th and U tend to concentrate in differentiates (ROGERS and ADAMS, 1969). Magmatic trends include also a positive correlation of radioelements with SiO_2 and K_2O and a negative correlation with CaO (DAWSON and GALE, 1970; PUTINSEV *et al.*, 1972; BULAKH *et al.*, 1973; DOSTAL *et al.*, 1975). In sediments, highest concentrations of uranium are frequently found in graywackes and argillaceous sandstones and lowest concentrations in deep-sea sediments (TUREKIAN and WEDEPOHL, 1961; HORN and ADAMS, 1966; BECKER *et al.*, 1972). In the Bahia granulites both thorium and uranium do not exhibit any of the magmatic trends discussed above, although the major concentrations of radioelements show a preference for Ca-poor salic granulites. The lack of any relationship of Th

Table 2. Concentrations of radioelements and heat production values in common metamorphic rocks and in shield areas.

	K(%)	U	Th	H.P.U.	N	
Epid.-amphi. facies		3.45	26.48			HEIER and ADAMS, 1965
Amphibolite facies		1.22	9.39			HEIER and ADAMS, 1965
Low-grade gran. f.		0.88	4.09			HEIER and ADAMS, 1965
High-grade gran. f.		0.39	0.93			HEIER and ADAMS, 1965
Migmatites	4.0	2.7	18.5	5.7	30	SMITHSON and DECKER, 1974
Migmatites	2.0	8.0	12.5	7.4	8	HUSMANN, 1956
Granite gneiss	3.0	1.7	14.5	4.3	99	LACHENBRUCH and BUNKER, 1971
Mica schist	2.7	4.0	14.3	5.4	7	SMITHSON and DECKER, 1974
Amphibolites	1.3	1.4	5.6	2.1	5	SMITHSON and DECKER, 1974
Granulites	2.71	0.70	13.34	3.32	31	This work
Canadial shield	2.58	2.45	10.3	4.55		SHAW, 1967
Australian shield	3.0	1.5	12.8	4.4		LAMBERT and HEIER, 1967
Baltic (crust as a whole)	1.5	1.9	5.7	2.94		GALDIN, 1974

N: number of samples.

and U with major-element chemistry may be related to the composition of the original material (heterogeneous nature, dominantly sedimentary character) but alternatively may be due to the behaviour of these elements during metamorphism.

Behaviour of Radioelements during Metamorphism

The fact that high-grade metamorphic terrains have usually lower U and Th contents than continental averages (see e.g. ADAMS *et al.*, 1959; HEIER and ROGERS, 1963; TAYLOR, 1964) is usually taken as the best proof that U and Th are mobile and become strongly depleted during regional metamorphism. Other findings, however, seem to disagree with this conclusion. PETROV *et al.* (1972) for example stated that radioelements behave as inert elements during metamorphism of sediments and that the observed variations in the U and Th and the Th/U ratios reflect the nature of the premetamorphic material. However, observations on the distribution of U and Th among mineral phases point out that these elements may be easily mobilized during metamorphism. For example, in magmatic rocks the distribution of uranium determined by alpha-particle autoradiography and fission-track methods show that U is concentrated mainly in small-volume accessory minerals; within the major minerals much of it is present in micro-inclusions. In both cases much of the α-emitting material is located at acid-leachable sites (HAMILTON, 1975). The behaviour of Th is less clear. Distribution of thorium in deuterically altered granites (BRIMHALL and ADAMS, 1969), as well as successful leaching experiments on monazite (BURGER *et al.*, 1967) favour a considerable mobility of this element under hydrothermal conditions. There are, however, many indications that Th is much less mobile than U. KILLEEN and HEIER (1975b) have recently observed in Norwegian Precambrian granites a relative movement of uranium with respect to thorium and attributed this to an event which has caused the oxidation of uranium. The environmental oxidative conditions may be, in fact, critical in determining a differential behaviour of uranium and thorium. Thus, oxidative conditions, together with possible site preferences of U and Th in minerals, are probably the main causes which determine a differential behaviour of uranium and thorium during metamorphism as frequently proposed (YERMOLAYEV and SOBORNOV, 1973). Rocks and fluids formed hydrothermally by contact metamorphism show very low Th/U ratios, in particular in the low-medium temperature hydrothermal processes (PLYUSHCHEV and RYABOVA, 1974). Thus it is highly probable that the extremely high Th/U ratios observed in the Bahia granulites were caused by a selective depletion of uranium in the metamorphic fluids. On the other hand, high Th/U ratios are rather common in most Archean metamorphic formations (LAMBERT and HEIER, 1967; HEIER and THORESEN, 1971) and were encountered also in Archean low-grade metamorphic rocks (ROGERS *et al.*, 1969). These findings were considered by FAHRIG and EADE (1968) as supporting the view that the continental crust about 3×10^9y ago had a Th/U ratio significantly higher than that of the whole earth represented by meteoritic material. The Amitsoq gneisses from Greenland dated about 3.8×10^9y have very low

U contents (*ca.* 0.6 ppm) and are high in non-radiogenic lead (MOORBATH *et al.*, 1975). This was obviously interpreted as indicating that the gneiss or their source region had undergone severe U depletion at or before *ca.* $3.7-3.8 \times 10^9$y (BLACK *et al.*, 1971; MOORBATH *et al.*, 1973). It is possible that such uranium depletions have frequently occurred in the Early Precambrian, thus leading to generalized high Th/U ratios. Pb isotopic data are of critical importance in testing this hypothesis.

HEAT PRODUCTION AND COMPOSITION OF THE CRUST IN DEEPLY-ERODED AREAS

The heat production resulting from the combination of all three elements in connection with heat flow may be of great importance in reconstructing the vertical compositional profile of a crust segment. While the heat production data for igneous rocks are capable of defining an igneous trend, estimates of heat production in various metamorphic rock types are less satisfactory because of small-scale variations in composition. Frequently, we observe some magmatic distribution trend such as heat production increase as the rocks become more felsic and approximate to granitic compositions (Table 2). It is also usually accepted that high-grade metamorphic rocks present the lowest heat production rate. The lowest continental layer comprising amphibolite-granulite facies rocks according to the model of SMITHSON and DECKER (1974) would be characterized by low heat production values in the range of 0.5–1.5 H.P.U.

Some researchers have observed a rather surprising linear relationship between heat flow and surface radioactive heat production at various sites of plutonic masses. The geological implications of these findings are far-reaching. Assuming a continental crust model (as that cited above) including a series of discrete layers each with constant heat production it can be concluded according to ROY *et al.* (1968) that the same erosional stability level was attained over large regions. LACHENBRUCH (1968), however, showed that an exponential dependence of heat source with depth is required for the correlation to hold for different erosion rates. For igneous masses such a condition may be satisfied admitting that the radioelements, as proposed in the model of ALBAREDE (1975), were depleted from low levels of the intrusion by migrating extensively in the fluids of the cooling magma and further in the cooling rocks. LACHENBRUCH (1970) proposed that the well documented (see also HAWKESWORTH, 1974) exponential distribution of heat sources with depth is achieved during tectogenesis by partial melting followed by upward movement and differentiation of the melt and fluids. Such processes are normally connected to high-grade metamorphism.

Heat flow is relatively uniform and low in Precambrian shields, possibly owing to the advanced depletion of heat sources from the upper mantle and/or to the high erosion rate of the most radioactive upper crust (POLYAK and SMIRNOV, 1968). Regarding heat sources, some authors (see e.g. WAKITA *et al.*, 1967; MORIOKA *et al.*, 1971) have postulated the existence of a generalized "barred" mantle residue of pyrolite from which crust-forming material have been extracted (RINGWOOD, 1962). One is tempted to ask: to what erosional level of the crust does an exposed granulite-facies terrain correspond and what is the true thickness and consequently the vertical compositional profile of the crust in such areas. From the foregoing it is clear that combined surface heat production-heat flow data in granulitic areas are of critical importance in studying these problems. Unfortunately, only few data are available and refer mainly to generalized shield areas and not to specific granulitic blocks. LAMBERT and HEIER (1967) compared Archean shield rocks of Australia with younger orogenic regions and found a much larger difference in the heat flow when compared with surface heat production. They hypothesize that the shield rocks are only representative of a very thin layer which rests on a crust further depleted in radioelements. The best opportunity for research on this problem would be afforded by examining a totally granulite facies crustal block like that of Bahia, although some ambiguity in conclusions may be inevitable on account of our imperfect knowledge of the crustal thermal gradient in Archean time. In any case, extremely important deductions could be drawn on the mode of rising to the surface of these originally deep-seated materials, i.e. whether they reflect simply very strong erosional rates of the crust, or, according to modern views of tectogenesis, they were brought to the surface through the intervention of horizontal or sub-horizontal (thrustings) crustal movements.

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